Aerosol-Cloud-Radiation Interactions in Atmospheric Forecast Models

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LONG-TERM GOALS

The long-term goal of this project is to gain a deep understanding of the role of atmospheric aerosols in affecting transmission of radiation through the atmosphere and in influencing cloud properties.

OBJECTIVES

The scientific objectives of this project are to identify the specific manner in which atmospheric aerosols determine cloud properties and to represent these interactions in atmospheric models. The technological objectives are to develop state-of-the-art instruments for aircraft sampling of aerosols that advance the long-term goals of the project.

APPROACH

The main technical approach is to conduct aircraft studies of the atmosphere, in which comprehensive sampling of atmospheric particles and radiative and cloud properties is carried out. The aircraft studies are complemented by laboratory investigations and theoretical analysis. Key individuals participating in this work are Professors John H. Seinfeld and Richard C. Flagan at the California Institute of Technology and Dr. Haf Jonsson at Naval Postgraduate School. Professor Seinfeld serves as Principal Investigator. Professor Flagan plays a key role in instrumentation development and planning of aircraft operations. As Chief Scientist of CIRPAS, Dr. Jonsson oversees all aspects of aircraft measurements and data management.

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14. ABSTRACT
The long-term goal of this project is to gain a deep understanding of the role of atmospheric aerosols in affecting transmission of radiation through the atmosphere and in influencing cloud properties.

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WORK COMPLETED

The MASE II field experiment was carried out in the eastern Pacific Ocean off Monterey, CA during July 2007 with the CIRPAS Twin Otter.   Comprehensive measurements were made of aerosol and cloud properties in areas both perturbed and unperturbed by local emissions.   Sixteen flights were made, with a total of 70 flight hours.   We are currently analyzing data from 13 individual clouds, 5 of which have evidence of ship tracks.   We are examining the ability to predict the cloud droplet size distribution observed.

RESULTS

1.   Rapid, Size-Resolved Aerosol Hygroscopic Growth Measurements: Differential Aerosol Sizing and Hygroscopicity Spectrometer Probe (DASH-SP) (Sorooshian et al., 2008)

We have developed a new instrument to perform rapid, size-resolved aerosol hygroscopicity measurements. The differential aerosol sizing and hygroscopicity spectrometer probe DASH-SP) employs differential mobility analysis in-concert with multiple humidification and optical sizing steps to determine dry optical size and hygroscopic growth factors for size-selected aerosols simultaneously at three elevated relative humidities. The DASH-SP has been designed especially for aircraft-based measurements, with time resolution as short as a few seconds. The minimum particle diameter detected with 50% efficiency in the optical particle counters (OPCs) is 135 ± 8 nm, while the maximum detectable particle diameter is in excess of 1 µm. An iterative data processing algorithm quantifies growth factors and "effective" refractive indices for humidified particles using an empirically derived three dimensional surface (OPC pulse height–refractive index–particle size), based on a calculated value of the "effective" dry particle refractive index. Excellent agreement is obtained between DASH-SP laboratory data and thermodynamic model predictions for growth factor dependence on relative humidity for various inorganic salts.   Growth factor data are also presented for several organic acids. Oxalic, malonic, glutaric, and glyoxylic acids grow gradually with increasing relative humidity up to 94%, while succinic and adipic acids show no growth. Airborne measurements of hygroscopic growth factors of ship exhaust aerosol during the 2007 Marine Stratus/Stratocumulus Experiment (MASE II) field campaign off the central coast of California are presented as the first report of the aircraft integration of the DASH-SP.

2. Aerosol Hygroscopicity in the Marine Atmosphere: A Closure Study using High-Resolution, Size-Resolved AMS and Multiple-RH DASH-SP Data (Hersey et al., 2008)

We have conducted the first closure study to couple high-resolution aerosol mass spectrometer (AMS) composition data with size-resolved, multiple-RH, high-time-resolution hygroscopic growth factor (*GF*) measurements from the differential aerosol sizing and hygroscopicity spectrometer probe (DASH-SP). These data were collected off the coast of Central California during seven of the 16 flights carried out during the MASE-II field campaign in July 2007. Two of the seven flights were conducted in airmasses that originated over the continental United States. These flights exhibited elevated organic volume fractions (*V F*$_{organic}$=0.46±0.22, as opposed to 0.24±0.18 for all other flights), 10 corresponding to significantly suppressed *GFs* at high RH (1.61±0.14 at 92% RH, as compared with 1.91±0.07 for all other flights), more moderate *GF* suppression at intermediate RH (1.53±0.10 at 85%, compared with 1.58±0.08 for all other flights, and no measurable *GF* suppression at low RH (1.31±0.06 at 74%, compared with 1.31±0.07 for all other flights). Organic loadings were slightly elevated in above-cloud aerosols, 15 as compared with below-cloud aerosols, and corresponded to a similar trend of significantly suppressed *GF* at high RH, but more moderate impacts at lower values of RH. A hygroscopic closure based on a volume-weighted mixing rule provided excellent agreement with DASH-SP measurements (R^2=0.79). Minimization of root mean square error between observations and predictions indicated mission-averaged organic 20 *GFs* of 1.20, 1.43, and 1.46 at 74, 85, and 92% RH, respectively. These values agree with previously reported values for water-soluble organics such as dicarboxylic and multifunctional acids, and correspond to a highly oxidized, presumably water-soluble, organic fraction (O:C=0.92±0.33). Finally, a backward stepwise linear regression revealed that, other than RH, the most important predictor for *GF* is *VF*$_{organic}$, indicating that a simple empirical model relating *GF*, RH, and the relative abundance of organic material can provide accurate predictions of hygroscopic growth in the marine atmosphere.

3. Comprehensive Simultaneous Shipboard and Airborne Characterization of Exhaust from a Modern Container Ship at Sea (Murphy et al., 2008)

We report the first joint shipboard and airborne study focused on the chemical composition and water-uptake behavior of particulate ship emissions. The study focuses on emissions from the main propulsion engine of a Post-Panamax class container ship cruising off the Central Coast of California and burning heavy fuel oil. Shipboard sampling included Micro-Orifice Uniform Deposit Impactors (MOUDI) with subsequent off-line analysis, whereas airborne measurements involved a number of real-time analyzers to characterize the plume aerosol, aged from a few seconds to over an hour. The mass ratio of particulate organic carbon to sulfate at the base of the ship stack was 0.23 0.03, and

increased to 0.30 ± 0.01 in the airborne exhaust plume, with the additional organic mass in the airborne plume being concentrated largely in particles below 100 nm in diameter. The organic to sulfate mass ratio in the exhaust aerosol remained constant during the first hour of plume dilution into the marine boundary layer. The mass spectrum of the organic fraction of the exhaust aerosol strongly resembles that of emissions from other diesel sources and appears to be predominantly hydrocarbon-like organic (HOA) material. Background aerosol which, based on air mass back trajectories, probably consisted of aged ship emissions and marine aerosol, contained a lower organic mass fraction than the fresh plume and had a much more oxidized organic component. A volume-weighted mixing rule is able to accurately predict hygroscopic growth factors in the background aerosol but measured and calculated growth factors do not agree for aerosols in the ship exhaust plume. Calculated CCN concentrations, at supersaturations ranging from 0.1% to 0.33%, agree well with measurements in the ship-exhaust plume. Using size-resolved chemical composition instead of bulk sub-micron composition has little effect on the predicted CCN concentrations because the cutoff diameter for CCN activation is larger than the diameter where the mass fraction of organic aerosol begins to increase significantly. The particle number emission factor estimated from this study is 1.3 x 1016 (kg fuel)-1, with less than one-tenth of the particles having diameters above 100 nm; 24% of the particles activate into cloud droplets at 0.3% supersaturation.

4.   On the Representation of Droplet Coalescence and Autoconversion: Evaluation using Ambient Cloud Droplet Size Distributions (Hsieh, et al., 2008)

In this study we evaluate eight autoconversion parameterizations against integration of the Kinetic Collection Equation (KCE) for cloud size distributions measured during the NASA CRYSTAL-FACE and CSTRIPE campaigns. KCE calculations are done using both the observed data and fits of these data to a gamma distribution function; it is found that the fitted distributions provide a good approximation for calculations of total coalescence, but not for autoconversion because of fitting errors near the drop-drizzle separation size. Parameterizations that explicitly compute autoconversion tend to be in better agreement with KCE, but are subject to substantial uncertainty, about an order of magnitude in autoconversion rate. Including turbulence effects on droplet collection increases autoconversion by about a factor of 1.82 and 1.24 for CRYSTAL-FACE and CSTRIPE clouds, respectively, and does not exceed a factor of   enhancement. Shifting the droplet-drizzle separation size from 20 to 25 μm results in about a twofold uncertainty in autoconversion rate. The polynomial approximation to the gravitation collection kernel used to develop parameterizations provides computation of autoconversion that agree to within 30%. Collectively, these uncertainties have an important impact on autoconversion, but are all within the factor of 10 uncertainty of autoconversion parameterizations. Incorporating KCE calculations in GCM simulations of aerosol-cloud interactions studies is computationally feasible by

using precalculated collection kernel tables, and can quantify the autoconversion uncertainty associated with application of parameterizations.

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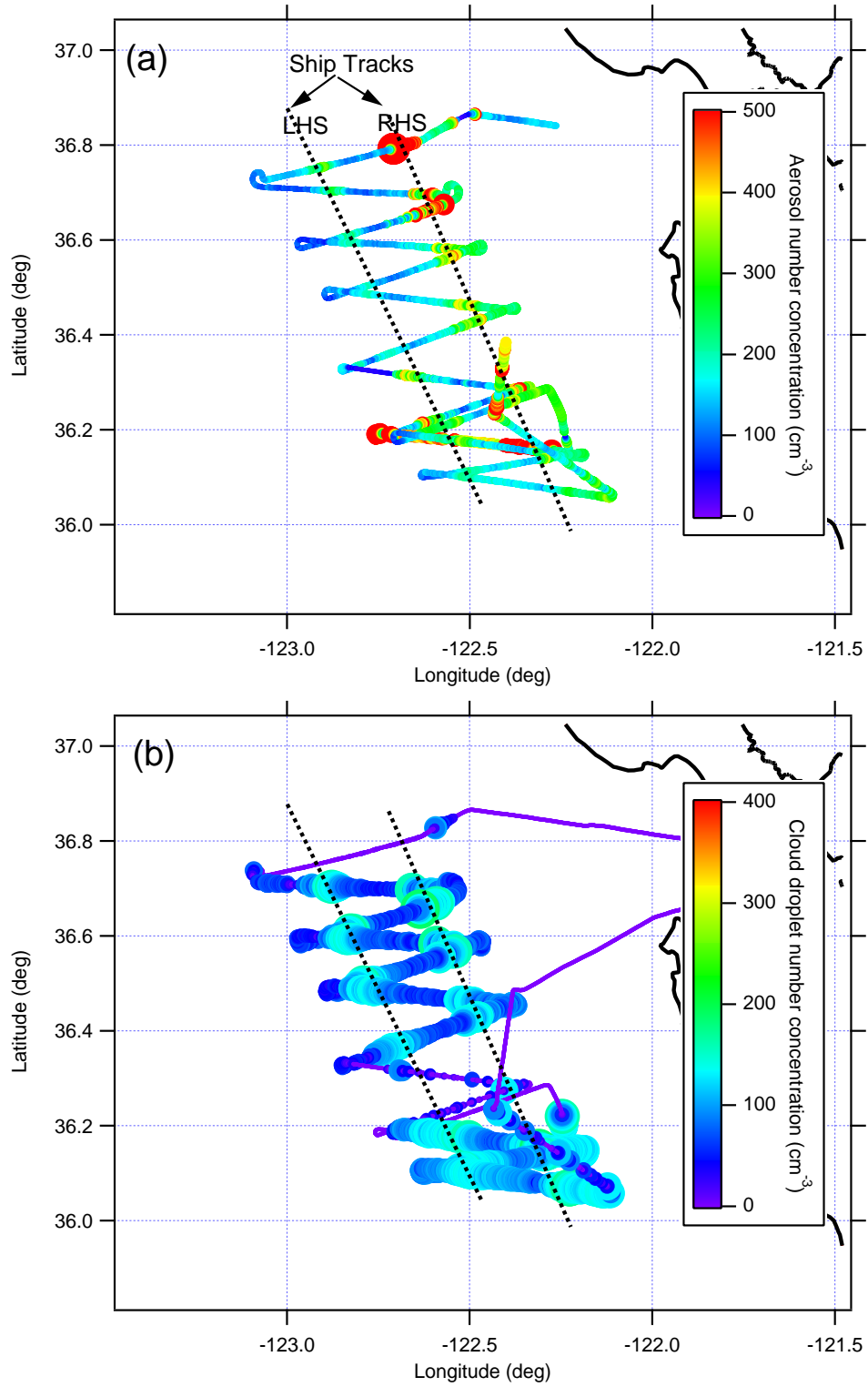
Figure 1. (a) Aerosol and (b) cloud droplet number concentration sampled on July 22, 2007 during the MASE II campaign. Ship tracks are clearly seen from the aerosol number concentration, denoted by two black lines (LHS and RHS for left-hand side and right-hand side, respectively); accompanied with the ship tracks are evidently two cloud lines. Total sampling period is about two hours.
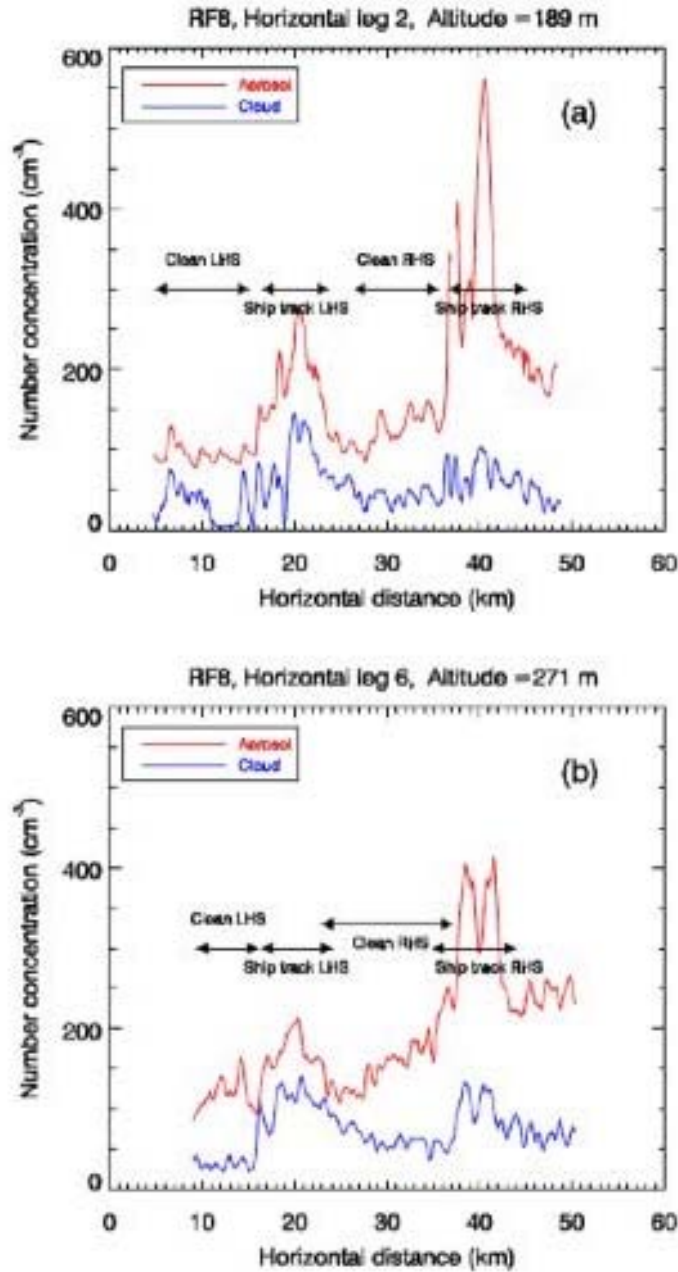
Figure 2. Cloud droplet and aerosol number concentrations from two horizontal flights: (a) from a leg in the lower and (b) upper portion of the cloud. From the spatial features of aerosol number concentration, the ship track and the relatively cleaner regions can be discerned (see arrows). The two ship tracks are present throughout the cloud, characterized by correspondingly higher cloud droplet number concentrations.